A Model of Coupled Heat and Moisture Transport in an Annular Clay Barrier

L. Rossi¹; H. I. Inyang²; J. Graham-Eagle³; and S. Pennell⁴

Abstract: The design of repository seals for deeply buried high-level radioactive wastes incorporates densely compacted clayey barriers around metallic waste canisters. In this paper, a mathematical model that is based on conservation of thermal energy and mass is developed to describe the locations of moisture and temperature fronts within a barrier, around a cylindrical waste canister of 1 m radius, containing radionuclides with half-lives that range from 100 to 10,000 years. The solution developed is axisymmetric: the moisture fraction w and temperature T vary only with time t, and radial distance r from the axis of the cylindrical waste canister. The model produces parabolic partial differential equations. The spatial domain is discretized such that ordinary differential equations that result are solved. Computations using a uniform mesh of 0.1 m and a cooling coefficient of $6.7 \times 10^{-2}$ with assumed but typical data on material properties, indicate that coupling of transport processes would be negligible in the case of radionuclides with long half-lives. Also, a quasisteady vaporization front can form and propagate outward over the course of several decades after waste emplacement. The evolution of the front is somewhat insensitive to the half-life used and the permeability of the clayey barrier material.

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Introduction

Some plans for long-term storage of high-level nuclear waste involve the burial of waste canisters within engineered clay barriers in geologically stable areas. The principal concerns with this storage plan are the potential for leaching of radionuclides into the surrounding groundwater and the associated risk of ecological and human exposure. Clay barriers can be altered by a variety of processes, including dehydration, infiltration of moisture and electrolytes, radiation damage, dissolution and leaching, precipitation, and illitization. To simulate these barrier degradation processes over long periods of time, it is crucial to be able to describe the fluid transport processes that deliver moisture and the substances that may contain to reaction sites in the barrier. A fundamental requirement for estimation of the rate of mineralogical changes in the barrier material during the service life of the repository is knowledge of the temperature and moisture content and gradient between the canister and the host rock. When the waste canister is first set in place, heat from the waste material causes a vaporization front to develop and move outward from the canister through the barrier, toward the host rock. As the waste decays and its heat output decreases, the vaporization front moves slowly back toward the canister. This paper focuses on the development of a model to describe the evolution of the vaporization front for use in calculating the temperature and moisture content of various regions of the barrier.

This paper is organized as follows. The significance of the transport processes that can alter the barrier over long periods of time is described in the following section. A mathematical model of the evolution and transport of thermal energy and moisture in the porous matrix surrounding the waste canister is developed in “Mathematical Model of Heat and Moisture Transport”. This coupled system of partial differential equations is adapted to a cylindrical geometry to quickly capture the essential configuration and coupled heat/mass transport pattern of the barrier. “Numerical Solution Algorithm” covers the implementation of a simple method-of-lines scheme to solve the system of partial differential equations in the cylindrical geometry. Sample computations of the advance of the vaporization front in an axisymmetric domain are presented in “Simulations”.

Relationship of Transport Processes to Barrier Deterioration and Contaminant Release

Establishment of hydraulic linkage between radionuclides contained in canisters and groundwater in a host rock is controlled by barrier alteration processes and the extent to which fluids can travel through the barrier. Barrier alteration and fluid flow processes in near-surface environments that are characterized by low confining pressures and alternating heat and moisture conditions have been described by several investigators among whom are Daniels et al. (2000), Daniels and Inyang (2003), Chen et al. (2000), Shi et al. (1999), Inyang et al. (1997), Bai et al. (1997), and Inyang et al. (2000).

¹Associate Professor, Dept. of Mathematical Sciences, Univ. of Delaware, Newark, DE 19716-2553.
²Duke Energy Distinguished Professor and Director, Global Institute for Energy and Environmental Systems (GIEES), Univ. of North Carolina at Charlotte, c/o C. C. Cameron Research Center, Room 238, 9201 University City Blvd., Charlotte, NC 28223-0001.
³Professor, Dept. of Mathematical Sciences, Univ. of Massachusetts Lowell, Lowell, MA 01854.
⁴Professor, Dept. of Mathematical Sciences, Univ. of Massachusetts Lowell, Lowell, MA 01854.

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In the deep subsurface environment in which pressures are very high and the temperature difference between the waste canister and the host rock can reach 150°C, coupled mass and heat transport will occur. Unlike the case of near-surface environments where barrier damage is mostly caused by alternating conditions such as wet–dry cycling, diurnal heating and cooling, and freeze–thaw action, the ingress of mineralized water that is typical of the deep subsurface into the emplaced barrier can induce fluid–barrier mineral reactions that alter the mineralogy of portions of the barrier layer. Indeed, mineralogical changes imply changes in the magnitude of barrier textural parameters such as radionuclide diffusion coefficient and permeability that may be significant with respect to the long-term release of radionuclides in the host rock and their subsequent migration into the near-surface environment. Earlier research has focused on characterization of the textural and hydraulic aspects of rocks that could impact the transport rates of radionuclides from emplaced wastes (Chambre and Pigford 1984; Bakhtar et al. 1993; Bai et al. 1996, 1997, 2000).

The potential for damage of clayey barriers by ionizing radiation emitted by packaged radioactive waste is much higher at the canister/barrier interface than at distant locations. The radiation level at the surface of waste canisters has been estimated as 100 mrem/h (Kehrmann et al. 1987). It is conceivable that in the event of canister damage, radiation levels within the barrier will be elevated at perforations if the waste is still in its highly radioactive state. The radiation component with the highest damage potential is gamma (γ) radiation. Then, its effect would mostly be restricted to the first few millimeters of barrier thickness around the canister. Clayey barrier materials possess γ-attenuation characteristics which may reduce gamma levels near the barrier’s external circumference to intensities that are incapable of generating activation-level dose rates. Data are unavailable on activation energies for radiolytic reactions in clays but related studies (Pusch et al. 1993) indicate that the exposure of montmorillonitic clay, saturated with weakly brackish water, to a γ-radiation dose of 3 x 10⁷ Gy at 130°C can cause the formation of minor amounts of quartz, CaSO₄, and MgSO₄. In their investigations, calcite (CaCO₃) and feldspars, which had been present as traces in the original clay, disappeared. In general, these changes are minor and were deemed insignificant with respect to overall barrier integrity by the investigators. In the case of water which may be present in the radiation field, the lifetime of excited molecules is below 10⁻¹¹ s (Bugaenko et al. 1993), perhaps too short for the initiation and completion of chemical reactions that can lead to significant textural changes in the barrier.

Under the expected high temperatures, at least during the initial radioactive decay phase of the waste, smectite crystals could dissolve in the inner annular section around the barrier if moisture requirements are met. Such moisture could be in the vapor phase and/or below saturation. Actually at emplacement, the barrier is expected to have very high density. Indeed experiments by Molecke and Torres (1983) show that 100% tamped bentonite (primarily Na-montmorillonite) has a density of 1.29 x 10⁻³ g/cm³, and data reviewed by Coulon et al. (1986) indicate that the residual saturation of clay at a standard pressure of 100 MPa ranges from 5.4 to 12.5%. Essentially, the barrier clay will be undersaturated initially. Significant increases in moisture content must occur through ingress from the surrounding rock.

The thermodynamics and kinetics of barrier alteration processes in the repository will be significantly influenced by the rate of moisture increase. This parameter itself is influenced by the nature of coupling of opposite processes of both heat and moisture. Although the ions that are needed to promote mineralogical transformations of barrier sections can be supplied through diffusion, increase in moisture (by convection), under the high environmental pressure of the repository environment, should be regarded as a significant mechanism of elevation of the moisture content of the barrier to levels required for illitization of the Na–montmorillonite. Smectite (a family of clays that includes montmorillonite) illitization requires the availability of potassium ions (K⁺). Feldspars and other minerals in the host rock can supply K⁺. The diffusion coefficients of ions through illite are generally higher than those through Na–montmorillonite. The implication is that the postconversion transport situation for radionuclides through the barrier (if they are released from the canister) may be higher than would be expected if barrier alteration were negligible. Diffusion of ions can be enhanced by increase in temperature. In their investigations of K⁺ diffusion in silty loam, Schaff and Skogley (1982) found that an increase in temperature from 5 to 30°C increased K⁺ diffusion between 1.6 and 1.7 times at each moisture level tested. K⁺ diffusion also increased with moisture content at constant temperature. Over the limited temperature range of 15–35°C, Youssef et al. (1961) studied the effects of temperature on the index properties of clayey soils and developed temperature-based correction factors for liquid limit and viscosity. From their data, an increase in temperature within the range they selected decreases both the liquid and plastic limits of soils. More recently, Kanno et al. (1995) have performed experiments that indicate that water diffusivity in compacted clayey soils (with a dry density of 1.8 x 10⁻³ g/cm³) increases significantly with temperature in the range of 25–60°C. The coupling of heat and mass transport (both moisture and ions) in clayey barriers of repositories is a significant determinant of barrier alteration processes and the potential transport rates of radionuclides from canisters in the long run.

Mathematical Model of Heat and Moisture Transport

A mathematical model is developed, to describe the moisture fraction or volumetric moisture content w, and temperature T, in the region surrounding a cylindrical high level nuclear waste canister buried deep underground. Immediately adjacent to the canister is an engineered clay barrier, and this engineered barrier is surrounded by a natural clay barrier as shown in Fig. 1.

Many of the essential mechanisms that determine the chemical environment near stored nuclear waste have already been identified and studied in short-term tests. Moisture is generated by condensation and lost through evaporation and vaporization. Moisture is transported by hydrodynamic diffusion. At higher-pressure gradients, inertial effects might become important but not in this regime. Thermal energy is generated by the stored nuclear waste.
and the condensation of water. Thermal energy is lost through vaporization. Finally, thermal energy is transported through thermal diffusion in the barrier particles and the intergranular pore moisture. Macaskill et al. (2001) studied a similar chemical system in stored bagasse in a different geometric configuration over shorter durations. In this analysis, the moisture fraction or volumetric moisture content, \( w \), is a dimensionless quantity equal to the ratio of the volume of liquid water to total volume; its value ranges from a minimum of 0 for a completely dehydrated region to a maximum of \( n \), the porosity of the matrix. In developing the model, the following assumptions are made.

1. The air pressure in the porous matrix is constant. This model is multiphase, but all regions that are not occupied by the matrix or by liquid water are assumed to be occupied by air. The vapor pressure in the air is determined such that it is in equilibrium with the far field moisture content at the far-field temperature. Any excess water vapor generated by evaporation is assumed to be transported out of the barrier region.

2. The water, air and porous matrix are in thermal equilibrium. This assumption is justified by the fact that the heat transport times between clay, air and water are on the order of seconds and minutes, whereas the hydrodynamic transport of moisture in clay requires days, months, and even years.

3. There are no open fractures in either the engineered barrier or the host rock because the engineered barrier will swell or the host rock because the engineered barrier will swell and close any fractures. Fractures in the host rock will be largely closed because of the very high overburden pressure that characterizes deep burial environments.

4. Mineralogical and chemical reactions within the barrier do not significantly affect the moisture and heat transport. Barrier alteration would result in a more complex coupled system of equations that will be treated in future work.

5. The hydraulic and thermal diffusivities are isotropic.

6. The solution is axisymmetric: that is, \( w \) and \( T \) vary only with time \( t \) and with radial distance \( r \) from the axis of the waste canister. The region of interest is a thick vertically uniform, annular pancake surrounding the storage cylinder as described by Carr and Basham (1983) and Seitz et al. (1987). The measurements of Kanno et al. (1995) support this assumption. This is not essential for the formulation of the basic model, but it permits a simple implementation of the model in a cylindrical geometry.

Conservation of thermal energy and water mass provides the two equations needed to solve for the two unknown quantities \( T \) and \( w \). The relevant processes for conservation of thermal energy density are thermal diffusion via Fourier’s law, evaporation and condensation as described in Eq. (1).

\[
(r_c + w p_w c_w) \frac{\partial T}{\partial t} - \nabla \cdot [K(w) \nabla T] = -\rho_o L_v (Z_w w e^{-L_v/RT} - Z_e V_{eq}) \tag{1}
\]

where \( \rho \) = density of clay; \( \rho_o \) = moisture density; \( c = \) heat capacity of the barrier; \( c_w = \) heat capacity of water; \( L_v = \) latent heat of vaporization of the water; \( Z_v = \) rate constant associated with the Arrhenius model for evaporation; \( R = \) universal gas constant; \( Z_e = \) condensation rate; and \( V_{eq} = \) equilibrium vapor pressure in the porous matrix which is assumed to be constant. The units of measurement of these parameters are provided in the Notation. The thermal diffusivity \( K(w) \) is given by Eq. (2).

\[
K(w) = k_b + w k_w \tag{2}
\]

where \( k_b = \) thermal conductivity of the barrier material; and \( k_w = \) thermal diffusivity of water. The equilibrium vapor content \( V_{eq} \) is determined from the far-field temperature \( (T_0) \) and moisture content \( (w_0) \) so that condensation rate is equal to the evaporation rate.

\[
V_{eq} = \frac{Z_w w_0 e^{-L_v/RT} - L_v/RT - Z_e V_{eq}}{Z_c} \tag{3}
\]

It is reasonable to neglect advection of thermal energy with the fluid flow because the fluid flux is much smaller than the thermal flux. Then, the porous matrix and fluid are in thermal equilibrium. The conductivity of air is neglected, as it is very small. The conductivity attributed to ambient vapor is disregarded because, under typical conditions, \( V_{eq} \) is on the order of one part in \( 10^4 \) and has no measurable effect relative to the conductivity of the barrier.

The relevant processes for conservation of water (moisture and vapor) are diffusion (driven by the moisture potential for unsaturated soils) and evaporation/vaporization.

\[
\frac{\partial w}{\partial t} - \nabla \cdot [D(w) \nabla w] = -Z_w w e^{-L_v/RT} + Z_e V_{eq} \tag{4}
\]

The hydraulic diffusivity \( D(w) \) incorporates a number of effects known in soil physics such as wetting and sorptivity. For this practical implementation, the diffusivity \( D(w) \) is fitted to measured data on Yolo light clay provided by Philip (1969, 1996).

\[
D(w) = 10^{-d(w)} \text{ m}^2/\text{s} \tag{5}
\]

where

\[
 d(w) = \begin{cases} 8.3 + 15000(w-0.03)^2, & w < 0.03 \\ 8.3 - 25171.7(w-0.1)(w-0.03)^2, & w \in [0.03,0.08025] \\ 7.0 + 25(w-0.4)^2, & w \in [0.08025,0.4] \\ 7.0 + (w-0.4)^2/4(w-0.3)(w-0.5)^2, & w \in [0.4,0.5] \end{cases} \tag{6}
\]

The results of the fit are shown in Fig. 2. Hydraulic diffusivity curves like the one in Fig. 2 have not been measured for many materials although the general behavior of the curve is somewhat universal for different clays.

The remainder of this paper is dedicated to the specific problem of quantifying the spatio-temporal location of the vaporization front in a cylindrical waste canister embedded in a clay barrier. One would expect any solution to Eqs. (1) and (4) with axisymmetric boundary and initial conditions to have axisymmetric solutions. The basic geometry is shown in Fig. 1.

To supplement Eqs. (1) and (4) it is necessary to determine the appropriate boundary conditions. The waste canister occupies the space \( 0 < r < R_0 \), the engineered clay barrier occupies the space \( R_0 < r < R_1 \), and the host rock occupies the space \( R_1 < r < R_2 \). Thus, the domain for the solution of the equations of the model is \( R_0 < r < R_2 \). The region occupied by the waste canister is treated as a “black box” save for a specified thermal flux \( H_0 e^{-\lambda t} \) where \( \lambda \) is the decay rate of the radioactive material in the waste canister from the edge at \( R_0 \), and it is assumed that the moisture in the clay barrier cannot penetrate the canister. Furthermore, it is assumed that the temperature and moisture fraction in the region surrounding the host rock have the constant values \( T_0 \) and \( w_0 \), respectively. Thus, the boundary conditions are...
Fig. 2. Hydraulic conductivity fit for Philip’s Yolo light clay data (Philip 1969, 1996)

\[
\frac{\partial T}{\partial r}(R_0,t) = -\frac{H_c e^{-\lambda t}}{2\pi R_0(k_b+wk_w)} \tag{7}
\]

\[
\frac{\partial w}{\partial r}(R_0,t) = 0 \tag{8}
\]

\[T(R_2,t) = T_0 \tag{9}\]

\[w(R_2,t) = w_0 \tag{10}\]

For initial conditions, it can be assumed that the temperature and moisture are equal to the ambient values throughout the system

\[T(r,0) = T_0 \tag{11}\]

\[w(r,0) = w_0 \tag{12}\]

Finally, the thermal flux and the hydraulic flux are continuous at the interface \(r=R_1\) between the engineered barrier and the host rock.

With a cylindrical domain, Eqs. (1) and (4) become

\[\frac{\partial T}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left[ rK(w) \frac{\partial T}{\partial r} \right] - H_N(T-T_0) - \rho_l L_v(Z_v w e^{-L_v/RT} - Z_v V_{eq}) \tag{13}\]

"Fig. 3. Graph of exact solution for steady state decoupled temperature distribution superimposed on computed results after letting simulation approach equilibrium (computations for \(H_N=0.002, H_c=5000, k_b=1, R_0=0.5, R_1=50, \) and \(T_0=20\)"

\[T = T_0 - \frac{H_c}{2\pi R_0 k_b} \left( \frac{1}{I_0\left( \sqrt{\frac{H_N}{k_b} R_0} \right)} + \frac{I_0\left( \sqrt{\frac{H_N}{k_b} R_1} \right)}{K_0\left( \sqrt{\frac{H_N}{k_b} R_1} \right)} \right) \times K_1\left( \sqrt{\frac{H_N}{k_b} R_0} \right) \left( I_0\left( \sqrt{\frac{H_N}{k_b} r} \right) - \frac{I_0\left( \sqrt{H_N/k_b R_1} \right)}{K_0\left( \sqrt{H_N/k_b R_1} \right)} \right) \times K_0\left( \sqrt{H_N/k_b r} \right) \tag{15}\]

A full three-dimensional model would include a term for the vertical diffusion of heat in Eq. (13) of the form \(D(w)\partial^2 T/\partial z^2\) plus far-field boundary conditions at the surface, but noting that the solution is essentially axisymmetric within a radial distance of many cylinder radii (Kanno et al. 1995), vertical heat diffusion can be replaced with a Newton cooling term. The Newton cooling term \(H_N(T-T_0)\) is a modification specific to the axisymmetric problem used to address physical and mathematical considerations. Physically, it is defensible to expect thermal energy to interact with surrounding material above and below the storage layer. Mathematically, there must be some form of heat loss to counteract the thermal energy input.

If moisture is absent from the system and the barrier has uniform thermal properties throughout, there is an exact, steady solution for temperature

\[T = T_0 - \frac{H_c}{2\pi R_0 k_b} \left( \frac{1}{I_0\left( \sqrt{\frac{H_N}{k_b} R_0} \right)} + \frac{I_0\left( \sqrt{\frac{H_N}{k_b} R_1} \right)}{K_0\left( \sqrt{\frac{H_N}{k_b} R_1} \right)} \right) \times K_1\left( \sqrt{\frac{H_N}{k_b} R_0} \right) \left( I_0\left( \sqrt{\frac{H_N}{k_b} r} \right) - \frac{I_0\left( \sqrt{H_N/k_b R_1} \right)}{K_0\left( \sqrt{H_N/k_b R_1} \right)} \right) \times K_0\left( \sqrt{H_N/k_b r} \right) \tag{15}\]
output of the waste canister, its surface temperature and the equilibriu
m temperature in the far field. Second, it provides a means of testing numerical
schemes by providing a reference solution for the decoupled problem.

**Numerical Solution Algorithm**

There are comprehensive large-scale computational packages for the
investigation of hydrogeological containment of nuclear waste. Specifically, Los Alamos National Laboratories has
developed FEHM (Zyvoloski et al. 2004), a finite element solver, and the Lawrence Berkeley Laboratory has developed TOUGH2 (Pruess 2004), a finite-difference solver. Both can perform time-
dependent simulations using models for multiphase flow through porous and fractured media. Both are large-scale codes for highly hetero-
egeneous environments. Since this work focuses on a simple model that isolates the small number of dominant effects in the
chemical environment near the waste storage cylinder, the approach taken here is to solve Eqs. (13) and (14) numerically using
the method of lines.

The method of lines is arguably one of the simplest schemes for attacking parabolic partial differential equations. Essentially,
the spatial domain is discretized, and the resulting coupled system of ordinary differential equations (ODEs) is solved using standard ODE solvers. In this case, the parabolic system is discretized to
second order using the method outlined by Skeel and Berzins (1990) and implemented in MATLAB. In Fig. 3, the numerical
approximation is shown to be in extremely close agreement with the exact solution [Eq. (15)] with sample parameter constants.

For the remainder of this paper, a uniform mesh of 0.1 m is used from the waste canister (with a 1 m diameter) out to a radius
of 2 m. From 2 to 3 m, a coarser mesh of 0.25 m is used. With this geometry, the cooling coefficient $H_N=6.7 \times 10^{-2}$ is chosen so
that with typical waste thermal outputs, the canister exterior reaches a temperature of roughly 250°C which is the upper re-

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**Fig. 5.** Prediction of vaporization front in Yolo light clay after core insertion with $\lambda=0.001$ year$^{-1}$; temperature is indicated with solid lines, and dimensionless moisture content is indicated with dashed lines.
gime of temperatures expected in repositories (with a factor of safety). Time integration is performed using a variable accuracy stiff solver with relative error tolerances set to be less than one part in $10^4$.

Simulations

This section presents a sample computation using typical and assumed but reasonable parameter values. These values are provided along with notations and units in the Notation. The graphs in Figs. 4 and 5 illustrate the development of the vaporization front following burial of the waste canister. Heat produced by the decay of the stored radionuclides raises the temperature of the barrier material and vaporizes barrier pore-water adjacent to the canister. A vaporization front moves outward from the canister, eventually reaching a state of quasiequilibrium. The canister’s heat output gradually decreases as its radioactive contents decay, causing the vaporization front to recede slowly back toward the canister. The details of the front’s evolution depend on the half-lives of the stored radionuclides and on the physical properties of the barrier material. The model presented in this paper can be used to study these effects, which in turn can have important implications for the possible chemical degradation of the barrier.

It is assumed that the porous matrix is almost saturated at $R_2$ so that $w_i = 0.40$. Half-lives for most of the radionuclides vary from 100 to 10,000 years. When half-lives for radionuclides exceed the warmup period, there is a separation of time scales so that advance of the front is independent of the half-life and the recession of the front scales proportionally to the half-life. In Fig. 4, it can be observed that for shorter lives (a), the decay timescale is coupled to the formation timescale so that the vaporization front advance is retarded compared to storage of materials with longer half-lives. For longer half-lives (b), the warmup time scale and the decay time scale are sufficiently separated such that the processes are decoupled. In Fig. 5, it is observed that the vaporization front forms within a century of introducing the waste. Since the half-life is 1 order of magnitude larger than the formation time, the decay of the stored waste will not have any significant impact on the formation or location of the front. Also, the relaxation time of the front will scale linearly with $1/\lambda$ as one would expect. The same results are observed for smaller values of $\lambda$. The simulation domain is longer than that shown ($R_2 = 50$ m). Thus, relaxation to equilibrium in the far field is fully established well beyond $r = 1.5$ m. Also a quasisteady vaporization front forms and propagates outward over the course of several decades. As the stored waste cools, the front retreats toward the core on the $1/\lambda$ time scale as one would expect. An example of this cooling regime is given in Fig. 6.

Perhaps the most important feature of these solutions is the presence of moisture behind the vaporization front. Thus, it is observed that between the waste and the vaporization front, there is a region where the temperature and moisture content may be high enough to support mineralogical transformations of the Na-smectite that is used as the primary constituent of the clay barrier. The transport of moisture into the porous matrix is computed from the moisture gradients in Fig. 7. As the vaporization front moves outward during the warmup period, the moisture transport profile moves outward as expected. Similar behavior is observed.

Fig. 6. Prediction of vaporization front in Yolo light clay as core cools with (a) $\lambda = 0.01$ year$^{-1}$ and (b) $\lambda = 0.001$ year$^{-1}$

Fig. 7. Computations of moisture flux in porous matrix for: (a) $\lambda = 0.01$ year$^{-1}$ and (b) $\lambda = 0.001$ year$^{-1}$
as the core decays and the front moves back toward the waste canister. In Fig. 8, information is provided on the temporal evolution of the temperature and moisture fraction at a fixed position close to the waste canister for two different values of λ. Simulations using permeabilities of twice and half those used in Figs. 4–8 indicate no appreciable change in the position of the vaporization front implying that the dominant mechanism is a balance between heat generation by the core and heat release through vaporization.

In summary, the sample computations show an initial propagation of a vaporization front away from the waste canister (the warmup phase) and the subsequent recession of the front back toward the canister as the stored radionuclides decay (the decay phase). The following physical effects are noted:

1. For radionuclides with half lives that exceed the warm-up period, the advance of the front is independent of the half-life;
2. For radionuclides with shorter half-lives, the advance of the front is retarded;
3. The rate of recession of the front is proportional to the half-life; and
4. Temperature and moisture content may be high enough to support mineralogical transformations of the clay barrier in the region between the front and the waste canister.

Conclusions

In this paper, a simple mathematical model that describes the temperature and moisture fraction in the region surrounding a nuclear waste canister has been developed and applied to a hypothetical case. The model incorporates thermal diffusion, diffusion of water (driven by the moisture potential for unsaturated soils), vaporization, and condensation. The partial differential equations that constitute the model are solved numerically using the method of lines. The results indicate that a vaporization front forms and initially moves away from the waste canister, reaching a location at which the effects of vaporization and inward diffusion of water are in balance. As the radioactive waste cools, the front slowly moves back toward the waste canister. The evolution of the front is relatively insensitive to changes in the half-life of the waste material and the permeability of the barrier material. A future paper will address the issue of possible chemical degradation of the barrier material through processes such as illitization.

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Notation

The following symbols are used in this paper:

\[ c = \text{heat capacity of barrier (10^3 J/mol K)}; \]
\[ c_w = \text{heat capacity of water (4.1819 \times 10^3 J/kg K)}; \]
\[ H_c = \text{thermal output of nuclear waste (2.2 \times 10^3 W)}; \]
\[ k_c = \text{heat conductivity of barrier (0.33 W/m K)}; \]
\[ k_w = \text{thermal conductivity of water (0.67 W/m K)}; \]
\[ L_v = \text{heat of vaporization of water (4.07 \times 10^3 J/kg)}; \]
\[ R = \text{universal gas constant (8.31 J/mol K)}; \]
\[ R_0 = \text{radius of storage vessel (0.5 m)}; \]
\[ T_0 = \text{ambient temperature underground (20°C)}; \]
\[ w = \text{moisture fraction ([0,0.5] for Yolo light clay)}; \]
\[ Z_c = \text{condensation rate (4.7 s}^{-1}); \]
\[ Z_e = \text{Arrhenius evaporation rate (3.41 \times 10^4 s}^{-1}); \]
\[ \rho = \text{density of porous substance (1.6 \times 10^3 kg/m}^3); \]
\[ \rho_w = \text{density of water (10^3 kg/m}^3). \]

References


